INCINERATED SEWAGE SLUDGE PRODUCTS AS AMENDMENTS FOR AGRICULTURAL SOILS: LEACHING AND PLANT UPTAKE OF TRACE ELEMENTS

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Abstract. Preliminary leaching column and greenhouse plant uptake studies were conducted in two soils with contrasting characteristics amended with varying rates (0 to 148.3 Mg ha^{-1}) of incinerated sewage sludge (ISS) and weathered sewage sludge (WISS) to estimate the leaching losses of trace elements from the soils amended with incinerated sewage sludge by products and to evaluate the uptake and accumulation of these elements in various parts of *Sorghum vulgaris* var. sudanense Hitche. ("Sorgrass"), a Sorghum-Sudan grass hybrid. Results of this study indicated that leaching of Cr, Cd, Zn, Cu, Ni, Fe and Mn from soils amended with ISS and WISS increased with increasing rates of amendment. Results of the leaching column study further revealed greater leaching losses from coarse-textured soil compared to medium-textured soil and also from ISS amended soils than with WISS amended soils. Results further suggested that the type of element and the interaction between the element and soil properties affected the leachability of various trace elements. The uptake study indicated uptake and accumulation of trace elements by plant parts increased with increasing rates of amendments. Greater plant uptake and accumulation of trace elements were observed in plant parts grown in soils amended with ISS compared to that of WISS. Results also indicated a greater accumulation of trace elements in below ground part of the plants (roots) compared to that was observed in above ground parts (shoots). Limited data obtained from this one season preliminary studies demonstrated that incinerated sewage sludge products from wastewater treatment plants could be used as soil amendments at low application (no more than 24.7 Mg ha^{-1}) for optimum plant growth, and dry matter yield without resulting in substantial accumulation of metals in plant parts at concentrations above the recommended critical limits and without causing significant leaching losses of various trace elements. It is imperative that long-term field studies are necessary to evaluate the long-term impact of using these new products in leaching and accumulation of various trace elements in plants and soils.

Keywords: incinerated sewage sludge, leaching, sewage sludge, trace elements, weathered incinerated sewage sludge.

1. Introduction

Purification and treatment of wastewater is one of the essential processes, which need to be accomplished by cities around the world. Solid waste, sewage sludge (SS), is a by-product produced by wastewater treatment plants during the purification process of wastewater. However, wastewater treatment plants in cities around the world encounter serious problems in disposing the SS due to lack of public acceptance because of their unpleasant odor, presence of excessive salt, acidity as well as the levels of some heavy metal in excess of critical limits. In general, SS is disposed in landfills (Basta, 2000; Power and Dick, 2000). Decrease in available landfill sites, increase in cost associated with landfill operation and the stringent regulations adapted by environmental protection agency (EPA) with respect to landfill operations (EPA 1985, 1993; McBride, 1995; NRC, 1996;) resulted in exploration of alternate disposal methods. These methods include production of ash (ISS) by incinerating dewatered activated sewage sludge and weathered incinerated ash (WISS) by dissolving and storing the incinerated by-product in ash ponds by wastewater treatment plants. These processes helped to overcome the problems associated with landfill operation and public acceptance of SS not only by removing unpleasant odor and reducing bulkiness by about 90% but also by reducing the cost of transportation to the field where it could be used as soil amendment.

Furthermore, the usability of the new products (ISS & WISS) as soil amendments in terms of leachability and bioavailability depends strongly on the so-called speciation of trace elements in to various chemical forms and their partitioning into various forms residues leaving the incinerator. Speciation and partitioning of trace elements in turn determined by the impact of operating conditions of (temperature and gas composition, residence time and the presence of reactive compounds such as chlorine, sulfur, or alumino-silicates) incinerator.

Compounds of trace elements with a high vapor pressure and a low boiling point enter the atmosphere easily and they are found mostly in the fly ash and flue gas from incineration of raw SS (Jakob *et al.,* 1995; Wey *et al.,* 1996). It is also of interest to note that presence of reactive compound such as chlorine has a strong influence on the trace element partitioning since all chlorides of trace elements generally have higher vapor pressure than that of their corresponding oxides. In addition, chlorine content in the combustion gas enhances the trace element volatilization because of the formation of volatile metallic chlorides (Belevi and Langmeier, 2000). Similarly, Morf *et al.* (2000) noticed that the transfer of Cu, Cd, and Pb into combustion flue gas increases with the sulphur load whereas Zn was not affected. It was also observed that, increased water content of the raw SS also has a reducing effect on the trace element transfer into the combustion flue gas (Verhulst *et al.*, 1996). Similarly the presence of excess mineral matter in the raw SS also affect the behavior of the trace elements through the formation of binary or tertiary oxides which can greatly reduce the volatilization and result in accumulation of these oxides in bottom ash (Jakob *et al.*, 1995; Verhulst *et al.*, 1996). In addition, incineration of raw SS substantially reduces the reactivity by destruction of organic compounds.

Multiple factors such as composition of the raw SS, operational conditions, and facilities available in the incinerator would definitely determine the physico-chemical properties of the new set of by-product (ISS) and which would be substantially different from that of raw SS. Furthermore, WISS is produced by the dissolution of ISS, which could lead possibly to more leaching losses in the waste ponds. The change in quantity and chemical status of various trace elements present in these new products (ISS & WISS) compared to that in raw SS would definitely alter the behavior of these elements when these new products are applied as soil amendments. Therefore, it is important to evaluate and compare the agronomic and environmental beneficial effects of these new by products (ISS & WISS) with unprocessed sewage sludge (SS), if one is going to use these new products as soil amendments.

In the past three decades of extensive research on unprocessed SS has helped to realize the beneficial effects of land application of SS in terms of increased crop production and improved soil quality (Page 1974; CAST, 1976; Chaney, 1994; Page and Chang, 1994; Basta, 1995; NRC, 1996; WEF, 1997; Power and Dick, 2000; Sajwan *et al.*, 2003). This resulted in renewed public interest in reusing and recycling of SS from wastewater treatment plants and dramatic increase in land application of SS from 20% to 54% in USA despite their unpleasant odor (Basta, 2000).

There is no literature currently available on the beneficial and safe use of these new by-products (ISS & WISS) derived from SS. Therefore, the evaluation of these new by-products alone or in combination with other by-products such as coal combustion by products (CCBP) is very important. This would provide valuable information about the potential impact of using these materials as soil amendments not only on crop production and soil fertility but also on environment. Therefore, the objectives of this study are to: (i) quantify leaching of trace elements from soils amended with varying rates of ISS & WISS and (ii) evaluate the uptake and accumulation of various trace elements in above and below ground plant parts of "Sorghum-sudan grass hybrid" grown under grown green house conditions. Result of this study would help to compare the merits of these new byproducts with the results obtained from previous experiments with unprocessed sewage sludge.

2. Materials and Methods

2.1. Leaching column study

Plexiglass columns, 32-cm long and 7-cm inner diameter, were used to study the leaching of various trace elements from soils amended with new by-products obtained by processing sewage sludge. Soils were dried, and sieved to pass a 2 mm sieve. A Whatman No. 42 filter paper was placed at the bottom of the leaching column and the soils were packed to a height of 30 cm in such away to attain their respective field level bulk density of $\sim$1.5 g cm^{-3}. A filter paper was placed on top of the soil surface and de-ionized water was applied at 1.5 mL min^{-1} flow rate using a peristaltic pump. Leachate was collected at one half-pore volume fractions (220 mL) at each leaching event.

This study was conducted by using two soil samples amended with ISS and WISS at the rates of 0, 24.7, 49.4, 98.8 and 148.2 Mg ha^{-1} (area basis & dry mass basis) to estimate the leaching of various trace elements. Selection of rates of amendments (ISS/WISS) in this study was based on previous experiences with our studies (Sajwan *et al.,* 1995; Alva *et al.,* 1999). Sajwan *et al.* (1995) showed a reduction in growth and yield of “Sorghum-Sudan grass hybrid” when the plants were grown in soil amended with mixture of sewage sludge and fly ash higher than 200 Mg ha^{-1}. It is very important to note that the chemical composition of sewage sludge material would vary according to the source and time of collection. Even though, the results obtained in these experiments has only limited value, they could provide some valuable information to estimate the potential leaching losses, uptake and accumulation of trace elements in various plant parts and to compare the results with unprocessed SS. The soil amendments (ISS & WISS) used in these studies were obtained from President street wastewater treatment plant located in Savannah, GA, USA. Some selected chemical properties of these by-products used in this study are presented in Table I along with the properties of unprocessed SS.

Leaching experiment was conducted with the fresh soil sample collected from the root zone of a Candler fine sand (sandy, hyperthermic, uncoated, Typic Quartzipsamments) collected from a citrus grove in Polk County, Lake Alfred, Florida and with fresh samples collected from the root zone of an Ogeechee sandy loam (Siliceous, thermic Typic Ochragult) from a field near Savannah State University, Chatham County, Savannah, Georgia. The soil in this horizon is more or less homogeneous within the root zone (top 30–35 cm depth) and this depth accounts for most (70 to 80%) of the root activities for the crops grown in these soils. Selected properties of the soil samples used in this study are also presented in Table I.

Triplicate soil columns were used to replicate each treatment level. Appropriate amount of amendment to represent each application rate was mixed with top 2.5 cm soil and repacked to attain the same bulk density as above to maintain original bulk density similar to that in the rest of the soil column. The amended soil columns were saturated with de-ionized water in an acid-washed bucket. Excess water was allowed to drain overnight. Leaching events were repeated at every 6d interval. Upon completion of each leaching event, soil columns were allowed to dry until the next leaching event (6d) at room temperature (30 °C). Leaching and drying conditions in this study were adapted to mimic the soil conditions in areas of Florida and southeastern Georgia during the summer months characterized by high temperature and frequent rainfall. The leaching/drying cycle was repeated 12 times for a total amount of leachate equivalent to 60 cm rainfall, which is about 48% of the mean annual rainfall (125 cm) of southeastern coast of Georgia. The electrical conductivity and pH of each leachate fraction were measured and ionic strength (in mM) was calculated using EC (μS cm^{-1}) values as per relationship of $I = 0.013$ EC (Griffin and Jurinak, 1973; Alva *et al.,* 1991).

TABLE I

Selected properties of unprocessed raw sewage sludge (SS), incinerated sewage sludge (ISS), weathered incinerated sewage sludge (WISS) materials and soils used in this study

Properties[a]	Units	SS[b]	ISS[b]	WISS[b]	Candler[c]	Ogeechee[d]
pH (1:1 water: soil)		6.00	8.00	7.70	6.80	5.50
EC	mS	3.36	1.59	0.23	0.07	0.05
C	%	39.50	1.05	1.10	0.50	1.20
N	%	4.57	0.15	0.02	0.04	0.06
S	%	1.11	0.20	0.01	0.01	0.02
P	mg kg^{-1}	13,250.00	2,912.00	274.50	1,130.00	206.00
Ca	mg kg^{-1}	15,980.00	13,250.00	7,150.00	369.20	3,284.30
Mg	mg kg^{-1}	1,655.00	1,632.00	1,110.00	562.40	262.20
Mn	mg kg^{-1}	191.20	139.50	83.50	14.50	14.10
Fe	mg kg^{-1}	10,420.00	9,250.00	6,200.00	591.40	661.50
Cu	mg kg^{-1}	219.50	210.00	24.00	38.40	1.20
Zn	mg kg^{-1}	542.500	526.00	69.50	92.60	7.40
Pb	mg kg^{-1}	37.10	29.50	10.50	20.90	4.90
Cd	mg kg^{-1}	4.10	1.30	0.40	0.40	0.60
Ni	mg kg^{-1}	16.60	14.30	8.50	nd	nd
Cr	mg kg^{-1}	27.40	17.50	8.00	7.40	4.30
Al	mg kg^{-1}	10,170.00	4,200.00	1,833.00	1,216.00	1,383.00
Sand	g kg^{-1}				967.00	860.00
Silt	g kg^{-1}				8.00	100.00
Clay	g kg^{-1}				25.00	40.00
Organic matter	g kg^{-1}				13.00	18.50
CEC	cmol kg^{-1}				2.20	3.90
Texture					Fine sand	Sandy loam

[a]Elemental compositions were determined by ICP-OES on acid-digest.
[b]Collected from President street wastewater treatment plant in Savannah, Georgia, USA.
[c]Collected from Polk County, Lake Alfred, Florida, USA.
[d]Collected from Chatham County, Savannah, Georgia, USA.

2.1.1. *Analysis of Trace Elements*

Concentrations of Cr, Zn, Cd, Cu, Ni, Fe and Mn in leachate were determined using inductively coupled plasma optical emission spectroscopy (ICP-OES; Plasma RL 3300, Perkin Elmer Inc., Norwalk, CT). ICP-OES instrument was calibrated with series of calibration standards (0, 0.2, 0.6, 1.0 & 2.0 mg L^{-1}) prepared in the same matrix (deionized water), which was used to leach the soil column. The analytical quality of entire set of samples was ensured by placing the same standards as samples in between every 15 samples. In addition, calibration standards were measured as samples to test the recovery of concerned trace elements in this study and whenever recovery was below 98%, instrument conditions were optimized and recalibrated to obtain at least 98% recovery of all the elements of interest. Furthermore, detection limits of Cr, Zn, Cd, Cu, Ni, Fe, and Mn are 2, 1, 1, 0.4, 5, 2 and 0.4 μg L^{-1} respectively. It is also of interest to report the detections limits of the ICP-OES used to analyze the samples obtained in this study, which would verify and validate the low values obtained during the analysis. Whenever the elemental concentration in samples exceeded the calibration standard, instrument was recalibrated with appropriate set of calibration standards and sample rerun was performed. The quantity of trace elements leached was calculated using the concentrations of each element and the volume of leachate fraction (220 mL) in each leaching event. Cumulative leaching loss of trace elements in this study was calculated, by summing up of loss of trace elements in each leaching event.

2.2. Plant uptake experiment with sorghum-sudan grass hybrid

Plant uptake experiment was conducted with fresh Candler fine sand and Ogeechee loamy sand and not with the soil samples used in leaching column study. The fresh soil samples were air dried and passed through a 2-mm sieve. This experiment consisted of five (treatments) application rates (0, 24.7, 49.4, 98.8 and 148.2 Mg ha^{-1}) of ISS or WISS, which were identical to the one used in leaching column study. These rates were applied to soil samples on a dry-weight basis to make a final weight of 3 kg and thoroughly mixed, and each rate was replicated three times. The bottom of each 4-kg polyethylene pot was lined with acid-washed gravel to facilitate smooth drainage, and then filled with 3 kg of amended soil. The acid-washing step was employed to clean the gravel, which would avoid or minimize any elemental contamination. Pots were incubated for two weeks at their respective field capacity moisture (7% and 14% for Candler fine sand and Ogeechee loamy sand respectively) and then planted with ten seeds of *Sorghum vulgaris* var. sudanense Hitchc. ("Sorgrass"), a Sorghum-Sudan grass hybrid. After two weeks, the pots were thinned to seven plants per pot and grown for 10 weeks. Pots were watered accordingly to maintain moisture content at their respective field capacity moisture throughout the growing period. The day and night temperatures of the greenhouse were maintained at 30 ± 3 °C and 24 ± 3 °C, respectively.

The above ground portion of each plant was harvested 10 weeks after planting, by clipping above the first node. The below ground portion of each plant was harvested carefully and processed for elemental analysis as described by Alva and Paramasivam (1998). However, this process is described very briefly. Harvested tissues from each pot were washed in detergent solution by rubbing with cheese cloth, rinsed in deionized water, soaked carefully in 10% HCl acid bath for about 30 seconds and followed by four successive rinses in deionized water baths to make sure that there were no soil particles adhering to (below ground part) roots. Then the washed plant parts were placed on brown paper sheets to dry at room temperature and then transferred to paper bags and placed in oven set at 70 °C for 72 h. The dried plant parts were weighed to the nearest 0.01 g, and ground in a Wiley mill to pass a

22 mesh (841 μm) sieve and stored in vials for further analysis. The ground plant samples were dry-ashed in a muffle furnace set at 550 °C for 6 h and then dissolved in 20 mL of 1 *M* HNO_3 (Council of Soil testing and Plant Analysis, 1980), and analyzed by ICP-OES for various trace elements as explained under analysis of leachate in leaching column study. Standard reference plant samples (from NIST) were used to check and confirm the analytical quality of ICP-OES.

2.3. Statistical analysis

Experimental data were analyzed using SAS (version. 8.1) as completely randomized design (CRD) with factorial treatment arrangement (2 Soil $\times$ 2 Sources of amendment $\times$5 rates of application) with three replicates per treatment combination. Means of cumulative trace elements (individual elements) leached were compared separately for soils by using Duncan's Multiple Range Test (DMRT) at 0.05 probability level. Similarly, means of dry matter yield and concentration of various trace elements in above and below ground parts of Sorghum-Sudan grass hybrid were compared by using Duncan's Multiple Range Test (DMRT) at 0.05 probability level.

3. Results and Discussion

3.1. Ionic strength of leachate

The measured electrical conductivity values EC (μS cm^{-1}) were converted into ionic strength (in mM) values using an established relationship of I = 0.013 EC (Griffin and Jurinak, 1973; Alva *et al.,* 1991). Ionic strength of the leachate is a direct measure of amount of ions present in the soil solution. Ionic strength of the leachate samples from soils amended with ISS was 4 to 6 times greater than that of the leachate samples obtained from the soils amended with WISS at all application rates irrespective of soil types used in this study (Figure 1). In general soils amended with WISS, ionic strength of the leachate reached peak value at one or one and a half pore volume (i.e., at second or third leaching event) irrespective of amendment rates and then gradually decreased below 2 mM for the sixth pore volume (twelfth fraction). It was quite evident in sandy soils (from Florida) amended with WISS showed the highest ionic strength value at the first leaching event (half pore volume) for all the amendment rates. The ionic strength values gradually decreased with subsequent leaching event and approached a stable and low value (~0.5 mM) irrespective of amendment rates in Candler fine sand by the sixth leaching event (Figure 1). In contrast, peak ionic strength value (3–4 mM) was observed at the second leaching (one pore volume) event in sandy loam soil sample from Georgia and then ionic strength values decreased irrespective of amendment rates. However, ionic strength values never dropped below 1 mM in sandy loam soil. The highest ionic strength

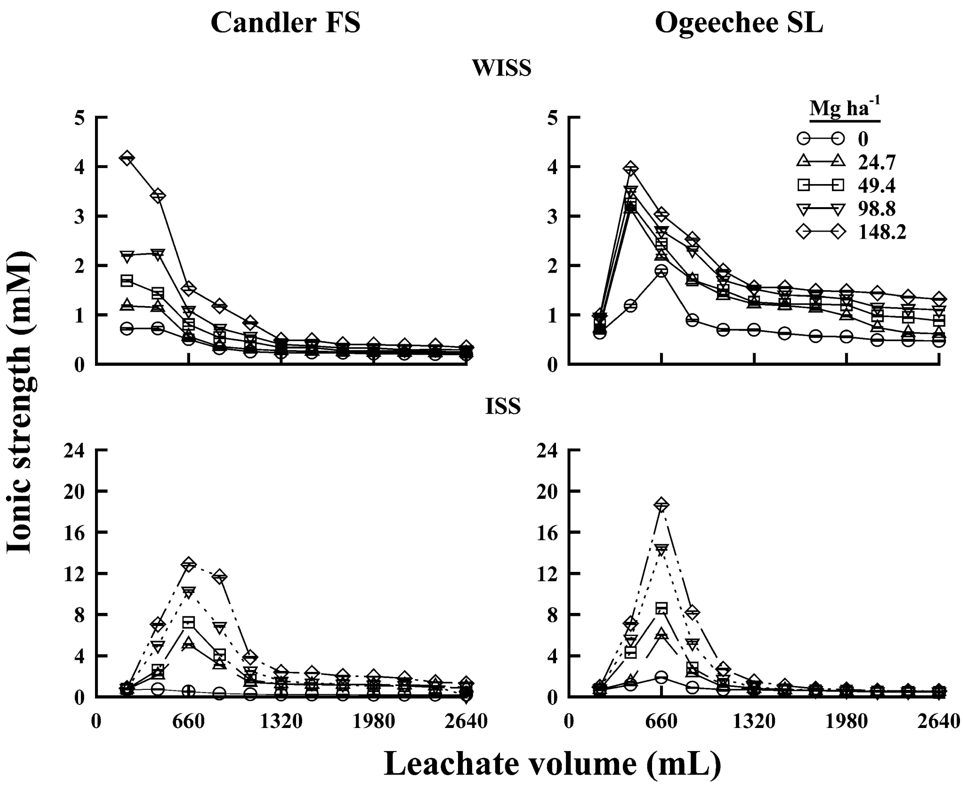

Figure 1. Ionic strength of leachate collected at various leaching events from soils amended with varying rates of ISS or WISS and unamended soils. Error bar on each point represents standard error of the means.

value observed with ISS amendment was about 20 mM [equal to 1540 μS cm^{-1} (1.54 mS cm^{-1}) electrical conductivity] while it was about 4 mM [equal to 310 μS cm^{-1} (0.31 mS cm^{-1})] with WISS amendment. Incidentally, these highest values are almost closer to the electrical conductivity values of the amendment types used in this study (Table I). The highest electrical conductivity value (0.31 mS cm^{-1}) observed in leachate from soil amended with highest rate of WISS amendment was very well within the limit ($<$0.7 mS cm^{-1}) we could normally expect with irrigation water. At the same time, the highest electrical conductivity value (1.59 mS cm^{-1}) observed in leachate from soil amended with highest rate of ISS was almost more than double the value we could normally expect with irrigation water. According to the guidelines for interpretation of water quality for irrigation (FAO, 1985), observed highest electrical conductivity value of the leachate collected from ISS amended soil at the highest rate (148.2 Mg ha^{-1}) fell below slight to moderate degree of restriction on use. Comparison of ionic strength data of the leachate from the same soil samples amended with SS at the rate of 74.1 Mg ha^{-1} from our previous study

(Alva *et al.,* 1999) indicated substantially greater values compared to the same soils amended with either ISS or WISS at the highest rate (148.2 Mg ha^{-1}) applied in this study. Even though the new by products used in this evaluation were also from municipal biosolids, they did not produce leachate with high electrical conductivity values even at the highest amendment rate (148.2 Mg ha^{-1}). This would be an added advantage for using new products derived from unprocessed sewage sludge, which has low levels of salt content. In contrast, unprocessed sewage sludge materials generally observed to have undesirable level of high salt contents (Sajwan *et al.,* 1995; Alva *et al.,* 1999; Basta, 2000).

3.2. Leachate pH

In soils amended with WISS, leachate pH did not vary significantly but was within the range of 6.3 to 6.6 throughout the study period (Table II). However, these values were relatively closer to the soil pH value of Candler fine sand (6.8). In contrast, pH values of leachate samples obtained from amended Ogeechee sandy loam were little bit higher than the pH value of the unameded Ogeechee sandy loam (5.5) used in this study (Table II). Similarly, in Ogeechee soil amended with ISS, the pH of the leachate increased to 6.6 on average irrespective of the rates of ISS application. However, there was a reduction in leachate pH observed in the first 3 to 4 leachate fractions then gradually stabilized and attained slightly higher (6.5) than the pH of the soil. Fluctuation in pH values could be due to the presence of various cations and anions in the system. We did not measure the composition of ions (cations & anions) and therefore we could not provide any possible explanation for the observed reduction in leachate pH since the pH of both ISS and WISS were greater than 7.0 (Table I and II). Furthermore, differences in buffering capacity of these soils, the ionic composition of leachate, and microbial decomposition process would help in explaining the observed pH changes. Similar trend was not observed in Candler fine sand with all ISS application rates and the average leachate pH was maintained between 7.0 and 7.2 (Table II).

3.3. Leaching of trace elements

In general, the amount of trace elements leached with 6 pore volumes of leachate indicated a greater leaching of trace elements in both soils amended with ISS compared to either WISS or unamended treatments used in this study (Table II). However, the leaching of Cd, Cr, and Ni from soils amended either with ISS or WISS at 148.2 Mg ha^{-1} was comparatively lower than the values obtained from the same soils amended with SS at 74.1 Mg ha^{-1} application rate (Alva *et al.*, 1999). This may be due to the concentrations of total Cd, Cr, and Ni determined in acid-digest by ICP-OES, which was relatively less in ISS, or WISS compared to that of SS samples (Table I). It is also important to note that trace elements (heavy metal) Cd and Hg would vaporize and disappear during incineration mainly as chloride

TABLE II

Comparison of mean leachate pH and total quantities of metals (μg) in 2,640 mL of leachate from Candler fine sand, or an Ogeechee sandy loam amended with various rates of weathered incinerated sewage sludge (WISS) or incinerated sewage sludge (ISS) materials and from unamended soils

Treatments Mg ha^{-1}	μg metals[a] in 2,640 mL leachate							
	pH	Cr	Zn	Cd	Cu	Ni	Fe	Mn
				Candler fine sand				
24.7 (WISS)	6.38 a	nd	81 g	nd	50 fg	nd	376 h	972 h
49.4 (WISS)	6.36 a	nd	92 g	nd	63 f	nd	680 g	1,504 g
98.8 (WISS)	6.44 a	nd	106 f	nd	120 e	nd	2,161 f	1,962 f
148.2 (WISS)	6.56 a	nd	233 e	nd	140 e	nd	3,748 e	2,923 e
24.7 (ISS)	7.19 a	38 c	2,164 d	18 b	4,448 d	48 d	14,047 d	5,715 d
49.4 (ISS)	7.11 a	46 bc	2,419 c	20 b	4,861 c	73 c	16,031 c	6,202 c
98.8 (ISS)	7.09 a	54 b	2,699 b	22 b	5,229 b	92 b	18,170 b	6,544 b
148.2 (ISS)	7.04 a	80 a	3,515 a	32 a	5,548 a	113 a	30,187 a	6,938 a
Unamended	6.94 a	nd	23 g	nd	37 g	nd	130 i	638 i
				Ogeechee sandy loam				
24.7 (WISS)	6.33 b	nd	29 g	nd	10 de	nd	9,865 g	1,123 g
49.4 (WISS)	6.43 b	2 d	38 g	nd	15 cd	nd	13,317 f	1,288 f
98.8 (WISS)	6.40 b	9 c	65 f	nd	19 c	nd	20,022 c	1,690 e
148.2 (WISS)	6.48 b	11 bc	117 e	nd	25 b	nd	23,646 b	2,015 d
24.7 (ISS)	6.59 b	6 c	275 d	nd	9 de	40 c	13,331 f	1,921 d
49.4 (ISS)	6.44 b	8 c	418 c	nd	12 d	48 bc	15,477 e	3,175 c
98.8 (ISS)	6.35 b	14 b	694 b	2 b	23 b	57 b	17,659 d	5,705 b
148.2 (ISS)	6.56 b	18 a	853 a	5 a	33 a	83 a	30,154 a	7,096 a
Unamended	5.33 a	nd	65 g	nd	7 e	nd	6,713 h	984.h

[a]Elemental compositions in leachate were determined by ICP-OES. Similar letters after a mean indicate no significant difference ($p < 0.05$) among treatment means within a column for each soil according to Duncan's Multiple Range Test. nd: non detectable.

(Abanades *et al.*, 2002). Similarly loss of Cu and Zn during incineration could be due to the amount of sulfur present and operating conditions in the incinerator (Morf *et al.*, 2000). In addition, process, which involves the preparation of WISS from ISS, would have significant impact on the content of various trace elements present in WISS. Therefore, leaching losses of trace elements from soils amended with ISS and WISS would be impacted by the inherent amount of trace elements present in them as well as the respective chemical forms of these trace elements.

In general, cumulative leaching of trace elements increased with increasing rates of amendments irrespective of the amendment or soil types used in this study. Leaching of most of the trace elements was greater in Candler fine sand compared to that from Ogeechee sandy loam for a given rate and source of amendment

(Table II). This is clearly an indication of the greater leaching ability for most trace elements in a coarse textured soil compared to that from medium textured sand. There was a significant difference between the two soils in the quantity of Cu and Zn recovered in the leachate and this might be due to the combination of higher amount of Cu and Zn in the incinerated sewage sludge (ISS) compared to that in WISS and textural differences between the soils used in this study (Table I and II). The differences in the concentrations of total Cu and Zn determined by acid digestion in both soil and amendment types are very obvious (Table I). Concentrations of total Cu and Zn were 32 and 16.5 times greater in Candler fine sand (38.4 & 72.6 mg kg^{-1} respectively) compared to that of Ogeechee loamy sand (1.2 & 4.4 mg kg^{-1} respectively). Furthermore, concentrations of total Cu and Zn were almost 8 times greater in ISS compared to that in WISS (Table I). About 60 to 70% of cumulative quantities of most metals leached were accounted for in the first 2 to 3 pore volumes (4 to 6 leaching events). Peak leaching of metals occurred at one and a half or 2-pore volume (3rd or 4th leaching event) irrespective of soils or amendments used (data not presented).

3.4. Average concentration of trace elements in leachate

Average concentration of various trace elements in leachate could be calculated by dividing the cumulative amount of any element leached for any soil/amendment type or treatment by total leachate volume (2.64 L) collected throughout the study period (Table II). The average concentration of Cr in the leachate from Candler fine sand amended with ISS varied in the range of 14.4 to 30.3 μg L^{-1} where as Cr was not detectable in the same soil amended with WISS at the same rates of amendment application (Table II). Leachate from Ogeechee soil amended with ISS and WISS showed Cr concentrations in the range of 0-to 6.8 μg L^{-1}. Similarly, the concentration of Cd in the leachate from Candler fine sand amended with ISS ranged from 6.8 to 12.1 μg L^{-1}, whereas Cd was not detectable in the leachate from the same soil amended with WISS. Similar non-detectable pattern of response was observed for the Ogeechee soil amended with ISS and WISS. The corresponding upper limits generally encountered in fresh water for Cr and Cd were 6 and 3 μg L^{-1} (Bowen, 1979). The average concentrations of Cr and Cd measured in leachate collected from both soils even at the lowest rate of amendment were substantially greater compared to that of the upper limits we generally encounter in fresh water. The critical upper limits for Cr and Cd for drinking water were 100.0 and 5.0 μg L^{-1} respectively (Rubenstein and Segal, 1993). However, the average concentration of Cd in the leachate from Candler fine sand amended with ISS exceeded the critical upper limit for Cd in drinking water even at the lowest amendment rate of 24.7 Mg ha^{-1}. However, it is of interest to note that these values are substantially lower than was generally reported as an average concentration range of Cd (0.02 to 130 μg L^{-1}) and Cr (0.5 to 1600 μg L^{-1}) from municipal landfill leachate monitored across various countries in Europe and USA (Baun and Christensen, 2004). This survey study was compiled by these authors from various studies reported in leachate samples collected from municipal landfills containing a mixture of household waste, commercial waste, industrial waste, and treatment sludges across Europe and USA (Baun and Christensen, 2004).

The average concentrations of Zn and Cu in the leachate from the Candler fine sand amended with 148.2 Mg ha^{-1} ISS were 1331 and 2102 μg L^{-1} respectively, whereas these values were 88 and 53 μg L^{-1} respectively for the Candler fine sand amended at the same rate of WISS. The corresponding concentrations for Ogeechee sandy loam amended with 148.2 Mg ha^{-1} ISS were 323 and 13 μg L^{-1} respectively, where as these values were 44 and 9 μg L^{-1} respectively for WISS amendment. The average concentration of Zn in fresh water was 15 μg L^{-1} (range 1 to 100) while that of Cu was 3 μg L^{-1} (range 0.2 to 30) respectively (Bowen 1979). Similarly, it is also of interest to note that the values generally reported as an average concentration range of Zn (0.05 to 155000 μg L^{-1}) from municipal landfill leachate monitored across various countries in Europe and USA (Baun and Christensen, 2004) was substantially less than that of the concentration observed in this study. In contrast, the average concentration of Cu observed in this study in Candler fine sand amended with ISS was substantially greater than the average concentration range (0.5 to 1300 μg L^{-1}) noted in survey study of Baun and Christensen (2004). Both ISS and WISS have substantial amount of Zn and Cu in it (Table I). Furthermore, Candler fine sand from Florida had substantially greater amount of Zn and Cu compared to that of Ogeechee loamy sand from Georgia (Table I). Elevated concentrations of Zn and Cu in the leachate from Candler fine sand amended with ISS or WISS came in part from the soil itself since this soil received periodic fungicide application for citrus cultivation. Results of this study suggest that, using ISS or WISS to amend the agricultural soils still could pose problem with respect to Zn and Cu loading into surface or ground water bodies. Elevated concentrations of Zn and Cu in water bodies not only depend on soil and amendment type but also depend on various physical, chemical and biological processes in those soils.

The average concentration of Ni in the leachate from the Candler fine sand amended with lowest to highest rates of ISS ranged from 18 to 43 μg L^{-1} where as the average concentration values of Ni were non-detectable for WISS amendment at the same range. In the case of the Ogeechee sandy loam the respective concentration ranges were 15 to 31 μg L^{-1} for ISS and non-detectable for WISS. The average concentration of Ni in fresh water generally encountered would be 0.5 μg L^{-1} and allowable range would be 0.02 to 27 (Bowen 1979). However, it is also of interest to note that these values are substantially lower than what was reported in a survey study as an average concentration range for Ni (1.0 to 3200 μg L^{-1}) from municipal landfill leachate collected and monitored across various countries in Europe and USA (Baun and Christensen, 2004).

The results of this leaching study suggest that amending soils with ISS even at the lowest rate could pose serious contamination problem of surface and ground waters. However, threat of contamination due to trace elements leaching and contamination of ground and surface water is substantially lower than that what was reported for landfill leachate samples collected and monitored in several European countries and in USA (Baun and Christensen, 2004). In general, concentrations of all metals increased with increasing rates of amendments. Therefore, it is advisable and safe to use the lowest amendment rate to avoid surface and groundwater pollution problem from these amendments.

3.5. Plant dry matter production

Dry matter production of above ground parts (shoots) increased with increasing rates of amendments irrespective of types of amendment. Irrespective of soil types, dry matter production of plants grown in soils amended with ISS at the highest rate was significantly greater compared to other amendment application rates. Dry matter production of above ground plant parts grown in Ogeechee sandy loam were greater than that of dry matter of plant parts grown on Candler fine sand (Table III). This was specifically true when amendment rate was 49.4 Mg ha^{-1} and above. Dry matter yield of below ground parts (roots) also followed similar trend of above ground parts (shoots) with respect to source and rates of amendments. However, dry matter yield of above ground part obtained at the highest rate of amendment with ISS was greater in plants grown on Ogeechee sandy loam soil and dry matter yield of below ground part was highest in Candler fine sand (Table III and IV). This might have been due to the fact that coarse textured soil (Candler fine sand) facilitated better root growth compared to that of medium textured soil.

3.6. Elemental uptake and accumulation by plant tissue

3.6.1. *Above Ground Plant Part (Shoot)*

In general, uptake and accumulation of most of the elements (metals) increased with increasing rates of amendments irrespective of sources of amendments and types of soils (Table III). Uptake of and accumulation of Cr, Cd, Cu and Pb in shoots were almost not detectable in both soils amended with WISS at all rates of application (Table III). Similarly, in soils amended with ISS uptake of Cr and Cd was not detectable, however, there was little uptake of Cu and Pb. Greater uptake of Cu and Pb was observed in Candler fine sand amended with ISS compared to that of ISS amended Ogeechee sandy loam. Also uptake of Ni and Mn was greater in Candler fine sand compared to Ogeechee sandy loam irrespective of type of amendments. However, this observation was opposite with respect to the uptake of Zn and Fe by above ground plant part (shoots) (Table III).

3.6.2. *Below Ground Plant Part (Roots)*

In general, uptake and accumulation of most of the elements (metals) increased with increasing rates of amendments irrespective of sources of amendments and types of soils (Table IV). However, no detectable uptake of Cd by roots in both soils amended

TABLE III

Comparison of dry matter yield and metal accumulation ($\mu g\ g^{-1}$) in above ground plant part (shoots) of Sudan sorghum grass grown in unamended Candler fine sand, or an Ogeechee sandy loamy and amended with various rates of either incinerated sewage sludge (ISS) or weathered incinerated sewage sludge (WISS) materials

Treatments Mg ha^{-1}	Dry weight (g)	[a]Metal concentration in above ground plant part (shoots) ($\mu g\ g^{-1}$)							
		Cr	Zn	Cd	Cu	Pb	Ni	Fe	Mn
			Candler fine sand						
24.7 (WISS)	3.9 e	nd	17 c	nd	nd	nd	138 e	26 d	42 f
49.4 (WISS)	5.5 d	nd	21 b	nd	nd	nd	173 c	36 c	97 e
98.8 (WISS)	9.0 c	nd	22 b	nd	nd	0.1 c	174 c	40 c	105 e
148.2 (WISS)	10.6 b	nd	26 ab	nd	0.2 d	0.4 c	177 c	52 b	138 d
24.7 (ISS)	3.6 e	nd	21 b	nd	1.0 c	0.4 c	158 d	35 c	243 c
49.4 (ISS)	5.7 d	nd	22 b	nd	2.6 b	0.9 b	170 c	44 c	246 c
98.8 (ISS)	10.2 b	nd	23 b	0.2 b	2.9 b	1.2 b	210 b	54 b	345 b
148.2 (ISS)	14.1 a	1.4 a	29 a	1.2 a	6.2 a	9.0 a	236 a	69 a	376 a
Unamended	1.1 f	nd	11 d	nd	nd	nd	17 f	19 d	29 f
			Ogeechee sandy loam						
24.7 (WISS)	3.0 f	nd	21.5 c	nd	nd	nd	99 e	32 d	26 f
49.4 (WISS)	7.0 e	nd	21.6 c	nd	nd	nd	102 e	37 d	33 f
98.8 (WISS)	15.7 b	nd	42.4 b	nd	nd	nd	106 e	44 c	189 d
148.2 (WISS)	15.9 b	nd	44.7 b	nd	nd	0.1 d	110 e	51 c	230 c
24.7 (ISS)	11.2 d	nd	37.0 c	nd	nd	nd	133 d	48 c	164 e
49.4 (ISS)	13.2 c	nd	41.1 b	nd	0.5 c	0.4 c	145 c	79 b	184 d
98.8 (ISS)	15.3 b	nd	48.7 a	nd	3.4 b	0.8 b	155 b	80 b	291 b
148.2 (ISS)	17.8 a	0.8 a	49.4 a	nd	4.5 a	1.4 a	165 a	94 a	345 a
Unamended	1.9 g	nd	8.9 d	nd	nd	nd	18 f	29 d	25 f

[a]Elemental compositions were determined by ICP-OES on dissolved dry-ashed plant samples in 1M HNO_3. Similar letters after a mean indicate no significant difference ($p < 0.05$) among treatment means within a column for each soil according to Duncan's Multiple Range Test. nd: non detectable.

with WISS at all rates, and also no detectable Cr uptake by roots at lower application rates of WISS in Candler fine sand (Table IV). Significant accumulation of metals was observed in roots compared to that of shoots in all treatments at all rates for both soils. This trend was observed for almost all the elements (metals) monitored in this study except Mn. Manganese accumulation was lower in roots at higher rates of amendments in both soils compared to that observed in shoots. In general, trace elements such as Zn, Mn, Ni and B are distributed uniformly throughout the plants. Copper and Cd generally accumulate in roots but some times in shoots. But Pb and Cr mostly accumulate in roots and very little in shoots (Wallace and Romney, 1972). However, in our investigation with Sorghum Sudan grass, most of

TABLE IV

Comparison of dry matter yield and metal accumulation (μg g^{-1}) in below ground plant part (roots) of Sudan sorghum grass grown in unamended Candler fine sand, or an Ogeechee sandy loam and amended with various rates of either incinerated sewage sludge (ISS) or weathered incinerated sewage sludge (WISS) materials

Treatments Mg ha^{-1}	Dry weight (g)	[a]Metal concentration in below ground plant part (roots) (μg g^{-1})							
		Cr	Zn	Cd	Cu	Pb	Ni	Fe	Mn
Candler fine sand									
24.7 (WISS)	0.6 e	nd	25 g	nd	9 f	0.8 f	92 e	437 f	43 f
49.4 (WISS)	0.6 e	nd	44 f	nd	32 de	5.1 de	107 d	485 e	64 e
98.8 (WISS)	1.1 d	0.2 d	60 e	nd	37 cd	7.1 cd	124 c	532 d	88 d
148.2 (WISS)	1.8 c	1.4 a	74 d	nd	57 b	9.0 c	134 c	627 c	206 b
24.7 (ISS)	0.5 e	0.5 c	83 d	0.2 b	27 e	3.5 e	125 c	350 g	67 e
49.4 (ISS)	1.2 d	0.9 b	97 c	0.2 b	42 c	8.9 c	180 b	656 b	161 c
98.8 (ISS)	3.3 b	0.9 b	113 b	0.2 b	53 b	12.8 b	217 a	1,008 a	164 c
148.2 (ISS)	4.6 a	1.5 a	156 a	2.9 a	67 a	18.9 a	231 a	1,022 a	244 a
Unamended	0.2 f	nd	18 g	nd	8 f	0.7 f	21 f	142 g	24 g
Ogeechee sandy loam									
24.7 (WISS)	0.4 f	2.1 f	82 g	nd	7 e	3.3 e	169 e	532 f	16 e
49.4 (WISS)	0.8 e	2.9 d	120 f	nd	10 de	3.7 e	186 e	795 e	29 e
98.8 (WISS)	1.9 c	3.0 d	240 d	nd	21 c	7.8 cd	263 d	965 d	137 d
148.2 (WISS)	2.0 c	4.5 b	272 c	nd	28 b	9.7 bc	282 d	1,046 c	219 b
24.7 (ISS)	1.6 d	2.5 e	147 e	0.3 c	10 de	4.0 e	277 d	969 d	134 d
49.4 (ISS)	1.8 c	3.3 c	238 d	0.6 b	13 d	6.0 cd	368 c	1,040 c	141 d
98.8 (ISS)	2.5 b	4.7 b	288 b	0.7 b	31 b	11.6 b	426 b	1,453 b	163 c
148.2 (ISS)	2.8 a	6.8 a	363 a	1.0 a	69 a	20.2 a	447 a	2,120 a	274 a
Unamended	0.2.f	1.4 f	48 h	nd	4 e	1.3 f	29 f	161 g	16 e

[a]Elemental compositions were determined by ICP-OES on dissolved dry-ashed plant samples in 1M HNO_3. Similar letters after a mean indicate no significant difference ($p < 0.05$) among treatment means within a column for each soil according to Duncan's Multiple Range Test. nd: non detectable.

the elements (metals) monitored accumulated in roots compared to that in shoots except Mn. The concentrations of Ni, and Pb in shoots observed in sorghum-Sudan grass grown in soils amended with ISS at higher rates were slightly greater than the average concentration of Ni and Pb observed in normal plants (Adriano, 2001). Similarly, the concentrations of Zn, Cd, Cu, Pb and Ni in roots observed in sorghum-Sudan grass grown in both soils amended with ISS at higher rates, were slightly greater than the average concentrations observed for these elements in normal plants (Adriano, 2001). The results of our study further indicate the potential for elements (metals) to accumulate in plant roots when ISS is used alone as land application for agricultural soils at higher rates. We therefore, suggest that care should be taken to avoid the use of higher rates of these by products to amend agricultural soils.

4. Conclusions

The results of this study indicate that leaching of most of the metals observed were comparatively lower with WISS than with ISS. The cumulative leaching of elements such as Cr, Cd, Pb and Ni range from nondetectable to 25 μg mL^{-1} in soils amended with WISS at all rates of amendment used in this study. Concentrations of most of the elements in the leachate were observed during early part of the leaching events at medium to higher rates (above 24.7 Mg ha^{-1}) of amendments, which exceeded the average concentrations of those respective elements found in natural fresh water environment. Greater cumulative leaching of elements was observed in coarse textured soil than in medium textured soil amended with both ISS and WISS.

In this study we observed a greater accumulation of Cr, Cd, Zn, Cu and Pb in plant roots when ISS was used as soil amendment at higher rates of application compared to that observed in plant shoots. Accumulation of the same elements was less in plants roots when WISS was used as soil amendments. Results of this study suggest that care should be taken in using these amendments to grow plants, which are consumed by, human and animal.

Over all,these amendments may be successfully used as soil amendments in agricultural lands at low application rates (less than 24.7 Mg ha^{-1}) for optimum plant growth without causing significant leaching. However, extensive in-situ studies in different soil types are suggested to draw comprehensive results, which could also help to monitor the transport of elements through soil. Also extensive study is recommended to evaluate their potential role and contribution to water pollution if they reach water body such as lake or groundwater reservoir.

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